Effects of atmospheric photo-oxidation on brown carbon aerosol spectral optical properties

Rajan Chakrabarty* and Benjamin Sumlin

Washington University in St. Louis, One Brookings Drive, St. Louis, MO, 63130, USA

**Presenting author (chakrabarty@wustl.edu)*

Light absorbing organic aerosols, optically defined as brown carbon (BrC), have been shown to strongly absorb short visible solar wavelengths and significantly impact earth's radiative energy balance. There currently exists a knowledge gap regarding the potential impacts of atmospheric processing on the optical properties of such particles. Climate models and satellite retrieval algorithms parameterize the optical properties of BrC aerosols as unchanging throughout their atmospheric lifecycle. Here, using integrated photoacoustic-nephelometer spectrometers, we investigated the effects of multiple-day photochemical oxidation on the spectral (375–1047 nm) optical properties of primary BrC aerosols emitted from boreal and Indonesian peatlands. Emitted particles were "aged" to timescales ranging from one to several days using an oxidation flow reactor under the combined influence of OH, O_3, and UV light. We found the largest effects of oxidation in the near-UV wavelengths, with the 375 nm imaginary refractive index (κ) and absorption coefficients of BrC particles decreasing by ~36% and 46%, respectively, and an increase in their single scattering albedo from 0.847 to 0.898. The spectral variability of κ follows the Kramers–Kronig dispersion relation for a damped harmonic oscillator. Direct radiative forcing efficiency calculations show the effects of aging on atmospheric warming attributed to BrC aerosols, which could be significant over snow and other bright surfaces.

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